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(54) Title: DISPERSIONS OF DELAMINATED PARTICLES IN POLYMER FOAMS (57) Abstract This invention relates to a polymer foam prepared by dispersing an organophilic multi-layered material into a melt comprising an olefinic or styrenic polymer having polar functionality so that at least a portion of the polymer intercalates between layers of the particles; and expanding the polymer with a blowing agent under such condition to form the polymer foam. The organophilic multi-layered material can also be dispersed into an olefinic or styrenic monomer, which can then be polymerized to form a polymer melt prior to, or along with the foaming step.		

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DISPERSIONS OF DELAMINATED PARTICLES IN POLYMER FOAMS

This invention relates to polymer foams prepared from resins having dispersed therein delaminated or exfoliated organophilic multi-layered particles.

Nanocomposites, which are compositions comprising single layers such as silicate
5 layers (1 to 100 nm thick) dispersed in a continuous polymer matrix, confer physical property enhancement to the polymer at a much lower ceramic content than conventionally processed glass- or mineral-reinforced polymers. These nanocomposites can be synthesized in a two-step process, with the first step being the modification of the multi-layered particles, generally by ionic exchange of sodium or potassium ions (which exist in natural forms of mica-type
10 silicates) with organic cations (for example, alkylammonium silicates or suitably functionalized organosilanes). This modification step can render normally hydrophilic mica-type silicates organophilic. In a second step, a melt-processible polymer (or a monomer which is polymerized *in situ*) and the intercalated particles can be compatibilized to form a polymer with enhanced physical properties.

15 Nanocomposite materials are described in U.S. Patents 4,739,007; 4,618,528; 4,528,235; 4,874,728; 4,889,885; 4,810,734; and 5,385,776; and in WO 93/11190.

The present invention addresses the need of improving the physical properties of polymer foams by providing a polymer foam that is prepared by a process comprising the steps of: a) dispersing organophilic multi-layered particles into a melt comprising an olefinic or
20 styrenic polymer having polar functionality so that at least a portion of the polymer intercalates between layers of the particles; and b) expanding the polymer with a blowing agent under such conditions to form the polymer foam.

In a second aspect, the present invention is a polymer foam prepared by a process comprising the steps of: a) dispersing organophilic multi-layered particles into a
25 polymerizable olefinic or styrenic monomer having polar functionality so that at least a portion of the monomer intercalates between layers of the particles; b) forming a polymer melt from the monomer; and c) expanding the polymer melt with a blowing agent under such conditions to form the polymer foam.

The polymer foam of the present invention can exhibit improved cushioning properties
30 and enhancement in elastic recovery properties. Also, improvement in insulation and ignition resistance, superior resistance to diffusion of liquids and of gases, and enhanced impact

strength can be realized in comparison to foams that are not prepared from the expansion of a suitably functionalized polymer intercalated between the layers of an organophilic multi-layered material.

The polymer foam of the present invention can be prepared by the steps of dispersing organophilic multi-layered particles into a melt comprising an olefinic or styrenic polymer having polar functionality so that at least a portion of the polymer intercalates between layers of the particles; and expanding the polymer with a blowing agent under such conditions to form the polymer foam. Alternatively, the organophilic multi-layered particles can be dispersed in an olefinic or styrenic monomer or prepolymer or a combination, and the monomer or prepolymer can then be polymerized *in situ* prior to or concurrent with the foaming step to form the polymer foam.

Organophilic multi-layered particles suitable for the preparation of the polymer foam can be prepared from hydrophilic phyllosilicates by methods well known in the art. Illustrative of such materials are smectite clay minerals such as montmorillonite, nontronite, beidellite, volkonskoite, hectorite, saponite, sauconite, magadiite, kenyaite, and vermiculite. Other useful multi-layered particles include illite minerals such as ledikite and admixtures of illites with the clay minerals named above. Other useful multi-layered particles, particularly useful with anionic polymers, are the layered double hydroxides such as $\text{Mg}_6\text{Al}_3(\text{OH})_{18.8}(\text{CO}_3)_{1.7}\text{H}_2\text{O}$ (see W.T. Reichle, *J. Catal.*, Vol. 94, p. 547 (1985), which have positively charged layers and exchangeable anions in the interlayer spaces. Other multi-layered particles having little or no charge on the layers may be useful in this invention provided they can be intercalated with swelling agents which expand their interlayer spacing. Such materials include chlorides such as ReCl_3 and FeOCl ; chalcogenides such as TiS_2 , MoS_2 , and MoS_3 ; cyanides such as $\text{Ni}(\text{CN})_2$; and oxides such as $\text{H}_2\text{Si}_2\text{O}_5$, V_5O_{13} , HTiNbO_5 , $\text{Cr}_{0.5}\text{V}_{0.5}\text{S}_2$, $\text{W}_{0.2}\text{V}_{2.8}\text{O}_7$, Cr_3O_8 , $\text{MoO}_3(\text{OH})_2$, $\text{VOPO}_4\cdot 2\text{H}_2\text{O}$, $\text{CaPO}_4\text{CH}_3\cdot \text{H}_2\text{O}$, $\text{MnHAsO}_4\cdot \text{H}_2\text{O}$, and $\text{Ag}_6\text{Mo}_{10}\text{O}_{33}$.

The hydrophilic multi-layered particle can be rendered organophilic by exchange of sodium, potassium, or calcium cations with a suitable material such as a water-soluble polymer, a quaternary ammonium salt, an amphoteric surface-active agent, and a choline compound, or the like. Representative examples of exchangeable water-soluble polymers include water-soluble polymers of vinyl alcohol (for example, poly(vinyl alcohol)), polyalkylene glycols such as polyethylene glycol, water-soluble cellulosic polymers such as

methyl cellulose and carboxymethyl cellulose, the polymers of ethylenically unsaturated carboxylic acids such as poly(acrylic acid) and their salts, and polyvinyl pyrrolidone.

Representative examples of the quaternary ammonium salts (cationic surface-active agents) which can be employed in this invention include the quaternary ammonium salts having octadecyl, hexadecyl, tetradecyl, or dodecyl groups; with preferred quaternary ammonium salts including dimethyl dihydrogenated tallow ammonium salt, octadecyl trimethyl ammonium salt, dioctadecyl dimethyl ammonium salt, hexadecyl trimethyl ammonium salt, dihexadecyl dimethyl ammonium salt, tetradecyl trimethyl ammonium salt, and ditetradecyl dimethyl ammonium salt.

Preferred organophilic multi-layered particles are those prepared by ion exchange of quaternary ammonium cations. A more preferred organophilic multi-layered material is a montmorillonite clay treated with a quaternary ammonium salt, most preferably dimethyl dihydrogenated tallow ammonium salt, commercially sold as CLAYTONE™ HY (a trademark of Southern Clay Products).

The organophilic multi-layered particles may also be prepared by the exchange of the sodium, potassium, or calcium cations with an inorganic material, a polymeric substance obtained by hydrolyzing a polymerizable metallic alcoholate such as Si(OR)_4 , Al(OR)_3 , Ge(OR)_4 , $\text{Si(OC}_2\text{H}_5)_4$, $\text{Si(OCH}_3)_4$, $\text{Ge(OC}_3\text{H}_7)_4$, or $\text{Ge(OC}_2\text{H}_5)_4$, either alone, or in any combination. Alternatively, the inorganic material can be a colloidal inorganic compound. Representative colloidal inorganic compounds which can be used include SiO_2 , Sb_2O_3 , Fe_2O_3 , Al_2O_3 , TiO_2 , ZrO_2 , and SnO_2 , alone, or in any combination.

The organophilic multi-layered material may also be prepared through exchange of functionalized organosilane compounds, as disclosed in WO 93/11190, pp. 9-21.

The concentration of the intercalated multi-layered particle in the polymer is application dependent, but is preferably not less than 0.1 weight percent, more preferably not less than 0.5 weight percent, and most preferably not less than 1 weight percent based on the total weight of the polymer; and preferably not greater than 40 weight percent, more preferably not greater than 20 weight percent, and most preferably not greater than 10 weight percent based on the total weight of the polymer.

The polymer foam is prepared, at least in part, from an olefinic or styrenic polymer having polar functionality. As used herein, the term "polar functionality" refers to a polymer

that contains a group or groups having a dipole moment, such as anhydride, carboxylic acid, hydroxy, silane, chlorine, bromine, or ester functionality, or combinations thereof. The polymer containing polar functionality can be prepared, for example, by polymerizing a polymerizable monomer containing polar functionality, by copolymerizing a polymerizable monomer containing polar functionality with another polymerizable monomer, or by grafting polar functionality onto a polymer backbone. The foamed dispersion may also be prepared from a polymer having polar functionality that is blended with a polymer that does not contain polar functionality.

Styrene or olefin polymers grafted with maleic anhydride or a vinyl silane, or styrene or olefin monomers copolymerized with maleic anhydride or vinyl silanes are especially suitable inasmuch as the maleic anhydride groups or silyl groups can serve to prevent re-agglomeration of the platelet particles. Preferred vinyl silanes include vinyl-Si(OR)₃ where each R is independently methyl, ethyl, or propyl, more preferably methyl or ethyl, and most preferably methyl.

Styrenic polymers such as those derived from styrene, α -methyl styrene, ethyl styrene, vinyl toluene, chlorostyrene and bromostyrene, and mixtures thereof, are also suitable. These styrene monomers may be copolymerized with other ethylenically unsaturated compounds such as acrylic acid, methacrylic acid, ethacrylic acid, maleic anhydride, maleic acid, itaconic acid, methyl acrylate, methyl methacrylate, butyl acrylate, butyl methacrylate, acrylonitrile, vinyl alcohol, amides, and butadiene.

Olefin polymers include polypropylene, polyethylene, and copolymers and blends thereof, as well as ethylene-propylene-diene terpolymers. Preferred olefinic polymers for the production of foam structures include polypropylene, linear high density polyethylene (HDPE), heterogeneously-branched linear low density polyethylene (LLDPE) such as DOWLEXTM polyethylene resin (a trademark of The Dow Chemical Company), heterogeneously-branched ultra low linear density polyethylene (ULDPE) such as ATTANETM ULDPE (a trademark of The Dow Chemical Company); homogeneously-branched, linear ethylene/ α -olefin copolymers such as TAFMERTM (a trademark of Mitsui PetroChemicals Company Limited) and EXACTTM (a trademark of Exxon Chemical Company), which can be prepared as described in U.S. Patent 3,645,922; homogeneously-branched, substantially linear ethylene/ α -olefin polymers such as AFFINITYTM polyolefin plastomers (a trademark of The Dow Chemical Company) and ENGAGETM polyolefin elastomers (a trademark of DuPont Dow Elastomers

LLC), which can be prepared as disclosed in U.S. Patents 5,272,236 and 5,278,272; and high pressure, free radical polymerized ethylene polymers and copolymers such as low density polyethylene (LDPE). The more preferred olefinic polymers are the homogeneously-branched linear and substantially linear ethylene copolymers with a density (measured in accordance with ASTM D-792) of 0.85 g/cm³ to 0.965 g/cm³, preferably from 0.86 g/cm³ to 0.92 g/cm³, more preferably from 0.86 g/cm³ to 0.90 g/cm³, and most preferably from 0.86 g/cm³ to 0.88 g/cm³, a weight average molecular weight to number average molecular weight ratio (M_w/M_n) from 1.5 to 3.0, a measured melt index (measured in accordance with ASTM D-1238 (190/2.16)) of 0.1 g/10 minutes to 100 g/10 minutes, and a melt flow ratio (I_{10}/I_2) of to 20 (measured in accordance with ASTM D-1238 (190/10)).

Preferred olefin polymers functionalized with a polar group include homogeneously-branched linear and substantially linear ethylene copolymers grafted with polar functionality, ethylene-acrylic acid (EAA) copolymers such as PRIMACOR™ (a trademark of The Dow Chemical Company), ethylene-methacrylic acid (EMAA) copolymers, and ethylene-vinyl acetate (EVA) copolymers such as ESCORENE™ polymers (a trademark of Exxon Chemical Company), and ELVAX™ (a trademark of E. I. du Pont de Nemours & Co.). More preferably, the olefin polymer functionalized with a polar group comprises a substantially linear ethylene-1-C₄-C₈ alkene copolymer grafted with maleic anhydride (the preparation of which is disclosed in U.S. Patent 5,346,963) or vinyl trimethoxysilane, most preferably an ethylene-1-octene copolymer grafted with maleic anhydride or vinyl trimethoxysilane and having a density of from 0.86 g/cm³ to 0.88 g/cm³.

Preferably, the foam is prepared from an admixture of an olefin polymer that contains polar functionality and an olefin polymer that is not functionalized. More preferably, the weight-to-weight ratio of the nonfunctionalized olefin polymer to the olefin polymer that contains polar functionality is not greater than 10:1, most preferably not greater than 5:1, and more preferably not less than 1:1, most preferably not less than 3:1.

Preferably, the concentration of the polar functional group on the polymer is not less than 0.1 weight percent, more preferably not less than 0.5 weight percent, and most preferably not less than 1 weight percent, based on the total weight of the styrene or olefin monomer(s) and the polar functional group. The preferred concentration of the polar functional group is not greater than 20 weight percent, more preferably not greater than 15 weight percent, more

preferably not greater than 10 weight percent, and most preferably not greater than 5 weight percent, based on the total weight of the polymer and the polar functional group.

Vulcanizable and thermoplastic rubbers used in the practice of this invention may also vary widely. Illustrative of such rubbers are brominated butyl rubber, chlorinated polyolefins
5 such as chlorinated polyethylene and chlorinated butyl rubber, fluoroelastomers, butadiene/acrylonitrile elastomers, silicone elastomers, poly(butadiene), poly(isobutylene), sulfonated ethylene-propylene-diene terpolymers, poly(chloroprene), poly(2,3-dimethyl-butadiene), poly(butadiene-pentadiene), chlorosulfonated polyolefins such as chlorosulfonated poly(ethylenes), and poly(sulfide) elastomers.

10 The organophilic multi-layered particles can be dispersed in *in situ* polymerized monomer or prepolymer by methods known in the art, for example, as disclosed in WO 93/11190, pp. 26-28.

The mixture may include various optional components which are additives commonly employed with polymers. Such optional components include nucleating agents, fillers,
15 plasticizers, impact modifiers, chain extenders, colorants, mold release lubricants, antistatic agents, pigments, and fire retardants. These optional components and appropriate amounts are well known to those of ordinary skill in the art.

The dispersed preformed polymer melt or the polymer prepared *in situ*, can be made into a foam by any suitable means, such as those known in the art. A chemical blowing agent
20 such as azodicarbonamide, 4,4'-oxybis(benzenesulfonyl hydrazide), or dinitrosopentamethylenetetramine, or a physical blowing agent such as carbon dioxide, CFCs, and light hydrocarbons, can be contacted with the polymer melt or reactive intermediates that contain the intercalated multi-layered particle to form a foamable gel, which can then be extruded through a die to form a foam product. The blowing agent may be incorporated into
25 the polymer melt or reactive intermediates by any suitable means such as extrusion, mixing, or blending. The blowing agent is preferably mixed with the melt polymer or reactive intermediates at an elevated pressure sufficient to disperse the blowing agent substantially homogeneously therein. Surprisingly, it has been discovered that the presence of the dispersed platelets can slow down the loss of incorporated gas from the foam, thereby providing a means
30 of maintaining the integrity of the foam.

Other methods of forming a foam are described in U.S. Patent 5,340,840, from column 13, line 49 to column 14, lines 1 to 35, along with the relevant teachings of U.S. Patents 4,323,528; 4,379,859; and 4,464,484. Also, see K. W. Suh and D. D. Webb in "Cellular Materials," from *Encyclopedia of Polymer Science and Engineering*, Vol. 3, Second Edition, pp. 1-59 (1985, and C. P. Park in "Polyolefin Foams" from *Handbook of Polymeric Foams and Foam Technology*, Hanser Publishers, New York, pp. 187-242 (1991).

The foams of the present invention exhibit useful properties, for example, enhanced yield strength and tensile modulus, even when exposed to polar media such as water or methanol; enhanced heat resistance; and enhanced impact strength as compared to the foams which do not include the dispersion layered materials. The improvements in properties are obtained even though smaller amounts of the layered material are employed. It has also been surprisingly discovered that the Asker C hardness of a 0.885 g/cm³ density polyolefin elastomeric resin (40 to 45) can be achieved, without substantially sacrificing resiliency or hysteresis of the 0.87 g/cm³ density resin, by adding small amounts (less than 5 parts per hundred) of an ion-exchanged organoclay to the 0.87 g/cm³ density resin grafted with a polar substituent, such as maleic anhydride or vinyl trimethoxy silane. Therefore, the desirable and unusual combination of high hardness, low hysteresis, and high resiliency can be achieved by the incorporation of the ion-exchanged organoclay material into a low density-foamed, polyolefinic material containing polar functionality.

The following examples are for illustrative purposes only, and are not meant to limit the scope of the invention.

Example 1 - VTMS-grafted Polyolefin Elastomer Bun Foams Containing Montmorillonite Organoclay

Vinyl trimethoxy silane (1.5 weight percent), AFFINITY™ EG 8200 resin (98.425 weight percent), and dicumyl peroxide (0.075 weight percent) were tumble blended for 30 minutes. The mixture was then compounded on a Werner-Pfleiderer ZSK-30 co-rotating twin-screw extruder that is operated under the conditions set forth in Table I. (AFFINITY™ EG 8200 resin is a substantially linear ethylene-1-octene copolymer having a melt index of 5 g/10 minutes, a density of 0.870 g/cm³, an I₁₀/I₂ of 7, and a M_w/M_n of 2.) Zones 1 to 5 refer to extruder heater zones.

Table I

Zone #1 (°C)	130
Zone #2 (°C)	172
Zone #3 (°C)	185
Zone #4 (°C)	211
Zone #5 (°C)	209
Die Temperature (°C)	197
Melt Temperature (°C)	225
Extruder Torque	70
Extruder RPM	100
Die Pressure (psi/kPa)	390/2690
Water Bath Temp (°C)	15

The resultant polymer (AFFINITY EG 8200.g.VTMS) was used to prepared a bun formulation as set forth in Table II. CLAYTONE™ HY (a trademark of Southern Clay Products) is a montmorillonite clay treated with dimethyl dihydrogenated tallow ammonium.

- 5 The coagent is SARTOMER™ 350 monomer, a trimethylolpropane trimethacrylate crosslinking coagent (a trademark of Sartomer Company). The formulation was prepared by mixing the components in a Banbury size BR internal batch mixer at 95°C to 110°C for 4 to 5 minutes. The mixture was then sheeted off into 1/4 inch (0.6 cm) to 1/2 inch (1.3 cm) thick sheets on a roll mill heated to 80°C. About 200 g of the above formulation was placed into a
- 10 chase with an opening of 5 inch x 5 inch x 0.5 inch (12.7 cm x 12.7 cm x 1.3 cm). The material was placed in a Tetrahedron MTP-8 compression press at 120°C for 20 minutes at 1 ton pressure (0.56 MPa), then immediately pressed in a Carver autohydraulic press (model 3895) at 175°C and 25 ton pressure (14 MPa) for 7 to 8 minutes. The material was then foamed at 150°C to 160°C, using 25 tons pressure (14 MPa) for about 45 minutes. Foam
- 15 density was measured by first cutting the foam into 1.5 inch x 1.5 inch squares (3.8 cm² x 3.8 cm²). The skin of the foam was then removed and the sample thickness adjusted to 0.5 inch (1.3 cm) using a Hobart slicer. Calipers were used to measure the dimensions of the squares. The sample was then weighed, and the density determined by dividing the mass by the volume. The static compression was determined by measuring the

initial thickness of the sample using calipers. The sample was then compressed 50 percent using 0.25 inch (0.63 cm) spacers in a static compression set jig as described in ASTM D-395 Method B. The foam density was $0.14 \text{ g/cm}^3 \pm 0.01$, the static compression was 62 percent ± 4 ; the Asker C hardness was 43 ± 2 , as measured by an Asker C durometer; the resiliency was 59 percent ± 1 , as measured in accordance with ASTM D-3574-86; and the hysteresis was 10.8 percent ± 0.1 .

Table II

Component	Amount (parts per hundred resin)
AFFINITY™ EG 8200.g.VTMS resin	100
Azodicarbonamide	3.0
Zinc Stearate	0.5
Zinc Oxide	1.0
IRGANOX™ 1010 antioxidant	0.1
Dicumyl Peroxide (40 percent active)	1.0
Coagent	1.25
Dibutyltin dilaureate	0.01
TiO ₂	3.0
CLAYTONE™ HY	4.5

Example 2 - VTMS-grafted Polyolefin Elastomer Bun Foams Containing Montmorillonite Organoclay

10 The procedure for forming the bun foams that was described in Example 1 was used in this example. The formulation is set forth in Table III.

Table III

Component	Amount (parts per hundred resin)
AFFINITY™ EG 8200.g.VTMS resin	100
Azodicarbonamide	3
Zinc Oxide	1
Dicumyl Peroxide	0.8
Coagent	2.5
TiO ₂	3
CLAYTONE™ HY	3

The compression set was 65.3 percent \pm 2.0 percent; the Asker C hardness was 51; and the resiliency was 58.

5 Example 3 - MAH-grafted Polyolefin Elastomer Bun Foams Containing Montmorillonite Organoclay

Maleic anhydride (MAH) grafted onto a substantially linear ethylene-1-octene copolymer was obtained from The Dow Chemical Company. (A procedure for preparing this material was disclosed in U.S. Patent 5,346,963.) The grafted material (AFFINITY™ EG 8200.g.MAH) contained 98.12 percent AFFINITY™ EG 8200, 1.4 percent MAH, 10 0.12 percent di-t-butyl peroxide, and 0.36 percent mineral oil. Bun foam formulations were prepared using the AFFINITY™ EG 8200.g.MAH and AFFINITY™ EG 8200. The formulation is set forth in Table IV. (IRGANOX™ is a trademark of Ciba-Geigy.)

Table IV

Component	Amount (parts per hundred resin)
AFFINITY™ EG 8200 resin	80
AFFINITY™ EG 8200.g.MAH resin	20
Azodicarbonamide	2.75
Zinc Stearate	0.5
Zinc Oxide	1.0
IRGANOX™ 1010 antioxidant	0.1
Dicumyl Peroxide (40 percent active)	1.5
Coagent	1.75
TiO ₂	3
CLAYTONE™ HY	4.5

A foam was prepared from the formulation as described in Example 1. The foam density was $0.14 \text{ g/cm}^3 \pm 0.01$; the Asker C hardness was 45 ± 2 ; the resiliency was 54 percent ± 1 ; and the static compression set was 80.

CLAIMS:

1. A polymer foam that is prepared by a process comprising the steps of:

a) dispersing an organophilic multi-layered material into a melt comprising an olefinic or styrenic polymer having polar functionality so that at least a portion of the polymer intercalates between layers of the particles; and b) expanding the polymer with a blowing agent under such conditions to form the polymer foam.

2. The polymer foam of Claim 1 wherein the olefinic or styrenic polymer having polar functionality contains anhydride, carboxylic acid, hydroxy, silane, chlorine, bromine, or ester functionality.

3. The polymer foam according to either Claim 1 or Claim 2 wherein the olefinic or styrenic polymer having polar functionality is an olefinic polymer or a styrenic polymer containing a maleic anhydride group or a vinyl-Si(OR)₃ group grafted thereto, wherein each R is independently methyl, ethyl, or propyl, and wherein the olefinic polymer is a polypropylene; a low density polyethylene, a linear high density polyethylene; a heterogeneously-branched, linear low density polyethylene; an ethylene-propylene-diene terpolymer; a heterogeneously-branched, ultra low linear density polyethylene; a homogeneously-branched, linear ethylene/ α -olefin copolymer; a homogeneously branched, substantially linear ethylene/ α -olefin polymer; a polystyrene; or a poly- α -methyl styrene.

4. The polymer foam as in any of Claims 1 to 3 wherein the organophilic multi-layered material is a quaternary ammonium salt of montmorillonite.

5. The polymer foam of Claim 4 wherein the quaternary ammonium salt is di-methyl di-hydrogenated tallow ammonium salt, octadecyl trimethyl ammonium salt, dioctadecyl dimethyl ammonium salt, hexadecyl trimethyl ammonium salt, dihexadecyl dimethyl ammonium salt, tetradecyl trimethyl ammonium salt, or ditetradecyl dimethyl ammonium salt.

6. The polymer foam as in any of Claims 1 to 5 wherein the melt further comprises an olefinic or styrenic polymer that does not contain polar functionality.

7. The polymer foam of Claim 6 wherein the olefin polymer that does not contain polar functionality is a homogeneously-branched, linear ethylene/ α -olefin copolymer and a

homogeneously-branched, substantially linear ethylene/ α -olefin polymer having a density of from 0.85 g/cm³ to 0.965 g/cm³, a M_w/M_n of 1.5 to 3.0, a melt index of 0.1 to 100 g/10 minutes, and an I_{10}/I_2 of from 6 to 20.

8. The polymer foam as in any of Claims 1 to 7 wherein the organophilic multi-layered particles comprise from 1 to 10 weight percent of the polymer melt, based on the weight of the resin.

9. The polymer foam of Claim 8 wherein the polymer melt comprises a mixture of a heterogeneously-branched, substantially linear ethylene-1-octene copolymer that is not functionalized with a polar group, and a heterogeneously-branched substantially linear ethylene-1-octene copolymer grafted with maleic anhydride or vinyl trimethoxysilane.

10. The polymer foam of Claim 9 wherein the polymer melt has a density of from 0.86 g/cm³ to 0.88 g/cm³.

11. The polymer foam of Claim 10 wherein the weight-to-weight ratio of the nonfunctionalized copolymer to the grafted copolymer is in the range of 1:1 to 10:1.

12. The polymer foam of Claim 11 wherein the weight-to-weight ratio of the nonfunctionalized copolymer to the grafted copolymer is in the range of 3:1 to 5:1.

13. The polymer foam as in any of Claims 1, 4, or 5 wherein the melt comprises an olefin polymer having polar functionality which is an ethylene-ethylene acrylic acid copolymer, an ethylene-ethylene vinyl acetate copolymer, or an ethylene-ethylene methacrylic acid copolymer.

14. A polymer foam prepared by a process comprising the steps of: a) dispersing organophilic multi-layered particles into a polymerizable olefinic or styrenic monomer having polar functionality so that at least a portion of the monomer intercalates between layers of the particles; b) forming a polymer melt from the monomer; and c) expanding the polymer melt with a blowing agent under such conditions to form the polymer foam.

INTERNATIONAL SEARCH REPORT

Inter. application No.

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A. CLASSIFICATION OF SUBJECT MATTER

IPC6: C08J 9/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: C08J, C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI, CA

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 9311190 A1 (ALLIED-SIGNAL INC.), 10 June 1993 (10.06.93) --	1-14
A	Dialog Information Services, File 351, Derwent WPI, WPI Accession No. 86-289269, Mitsubishi Petrochemical Co Ltd: "Glass fibre-reinforced composite resin sheet - comprises short glass fibre contg. thermoplastic sheet laminated to foamed thermoplastic res sheet contg. micropowdered inorganic filler", &JP,A,61213140, 19860922, JP,A,8553568, 19850318, 198644 B --	1-14
A	US 4889885 A (ARIMITSU USUKI ET AL), 26 December 1989 (26.12.89) --	1-14

☒ Further documents are listed in the continuation of Box C.☒ See patent family annex.

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- *P* document published prior to the international filing date but later than the priority date claimed

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X document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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Date of the actual completion of the international search

3 June 1997

Date of mailing of the international search report

0 5. 08. 97

Name and mailing address of the International Searching Authority



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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US 97/02644

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5340840 A (CHUNG P. PARK ET AL), 23 August 1994 (23.08.94) -----	1-14

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT
Information on patent family members

01/07/97

International application No.
PCT/US 97/02644

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